Tritium decontamination of machine components and walls

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Tritium decontamination techniques for machine components and their application at tritium handling facilities are reviewed. These include commonly used methods such as vacuuming, purging, thermal desorption and isotopic exchange as well as less common methods such as chemical/electrochemical etching, plasma discharge cleaning, and destructive methods. Problems associated with tritium contamination of walls and use of protective coatings are reviewed. Tritium decontamination considerations at fusion facilities are discussed.

1. Introduction

The general objectives of tritium decontamination of systems and components are:

- to reduce tritium hazards to sufficiently low levels to permit maintenance to be safely carried out at minimum dose;
- to clean up components for repair, modifications, reuse or shipment off-site;
- to minimize system tritium inventory, tritium outgassing and contamination spread from equipment and surfaces; and
- to reduce the tritium levels in waste to meet disposal criteria.

The unique feature of tritium contamination is the very high mobility of tritium. It will readily absorb into many materials and then subsequently outgas giving rise to chronic airborne contamination. Tritium contamination can readily spread from one area or object to another. It can diffuse through many materials resulting in contamination of the bulk as well as on the surface. It can be converted from one chemical form to another; the most common and important example is conversion of tritium gas (HT) to the radiologically much more hazardous tritium oxide (HTO). The principal modes of occupational exposure to tritium are inhalation of airborne tritium, diffusion of airborne HTO through skin, and uptake of tritium through skin contact with tritium-contaminated surfaces.

The choice of a decontamination method depends on the objective of the decontamination, the nature, size and configuration of the material and surface being decontaminated (metals, organics, ceramics, concrete), the chemical form of tritium (HT, HTO, organically bound), whether the tritium is largely confined to the surface or has permeated through the bulk, and whether decontamination has to be carried out in situ.

Tritium contamination of containment structures such as concrete walls and confinement rooms such as caissons gives rise to tritium out-gassing problems. During facility decommissioning, tritium contaminated concrete and structural materials may present large volumes of low level waste.

2. Standards and criteria

2.1. Surface contamination

Surface contamination is usually measured by the surface smear method (swipe) and expressed in units of Bq/m^2, $\mu Ci/m^2$ or dpm per 100 cm^2 where dpm are disintegrations per minute. The 100 cm^2 represents the averaging area in one swipe measurement. Somewhat different criteria have been used by different countries and institutions.

For unconditional release off-site, the surface contamination limit of 1 $\mu Ci/m^2$ (37 kBq/m^2) has been adopted in countries and institutions such as France, Canada (Ontario Hydro), and the Japan Atomic Energy Research Institute [1]. This value is conservative

since it is the same as that for other beta-gamma emitters.

In the U.S., present DOE (Department of Energy) surface contamination limits [2] for beta-gamma emitters apply to tritium since there are no separate limits for tritium. These limits are: 10^3 dpm per 100 cm^2 (1.7 kBq/m^2) removable activity and 5×10^3 dpm per 100 cm^2 (8.3 kBq/m^2) for fixed plus removable activity. Components with contamination below these levels may be released from controlled areas. Proposed surface radioactivity guides will increase these limits to 10^5 dpm per 100 cm^2 (0.17 MBq/m^2) for all forms of tritium.

For occupational exposure control, the ICRP [3] suggested a surface contamination limit of 10 μCi/m^2 for working with beta-gamma emitters and that for tritium this limit may be relaxed by a factor of 100, i.e. to 1000 μCi/m^2.

In Ontario Hydro, Canada, Derived Surface Concentrations (DSCs) for occupational exposure control have been developed for HTO and HT [4]. The DSC for HTO is 0.37 GBq/m^2 and corresponds to a potential annual whole body exposure of 5 rem. The DSC for HT is 18 MBq/m^2 corresponding to the annual skin dose limit of 50 rem. These dose limits are for occupational exposure. The DSC for HT is 20 times more restrictive than that for HTO. This is conservatively based on the uptake of tritium through skin contact with HT contaminated metal surfaces and subsequent high local dose to skin. No special precautions are required at contamination levels less than 1 DSC. Between 1–50 DSC, local ventilation control is required. Working with levels above 250 DSC requires close health physics supervision.

2.2. Waste disposal criteria

ANDRA, the organization for waste storage in France, accepts tritiated wastes, in 200 L drums, if the outgassing rate is less than 0.1 MBq (3 μCi) per day. The shallow land burial site Centre de la Manche accepts tritium wastes less than 74 Mq/kg (2 Ci/t) [5].

In Germany, the Konrad waste disposal site puts a tritium content limit of 470–960 GBq (13–26 Ci) per package and a tritium outgassing limit of 1.5 GBq (0.04 Ci) per year per package [6]. The tritium outgassing rate limits proposed for the Gorleben site ranges from 1.1–110 MBq (0.03–3 mCi) per year per package for disposal in tunnels and 190–1900 MBq (5–50 mCi) per year per package for bore holes [6].

The U.S. Nuclear Regulatory Commission 'Class A' near-surface disposal limit for tritium is 1.5 TBq/m^3

(40 Ci/m^3) [7]. Separate values for tritium are not given for 'Class B' or 'Class C'.

The Japan Atomic Energy Research Institute [1] classifies solid tritium waste containing less than 3.7 GBq (0.1 Ci) per container as low level waste.

3. Tritium decontamination techniques

Before a decontamination operation, the material surface should be characterized if possible and the surface tritium contamination level determined, typically by the 'swipe' method. The tritium outgassing rate may also be an important and useful indication of contamination level. Knowledge of the history of the material (operating conditions such as tritium pressure, temperature and time of contact with tritium, chemical form of tritium, etc.) may give indication as to whether the tritium is mainly present at the surface or has largely permeated through the bulk. Broadly, the following techniques, in various combinations, have been used for tritium decontamination:
(a) superficial cleaning and washing;
(b) vacuuming, purging;
(c) thermal desorption;
(d) isotopic exchange;
(e) chemical or electrochemical etching;
(f) plasma discharge methods;
(g) destructive techniques such as melting.

3.1. Washing, purging, thermal desorption and isotopic exchange

Surface decontamination by scrubbing with sandpaper followed by emery cloth polish, and by washing (with or without detergents) removes tritium-contaminated dust, dirt, oil, grease, etc. as well as loosely bound tritium. The process is slow, tedious and tends to generate low activity wastes. Tritium regrowth due to permeation from the bulk to the surface following surface decontamination at moderate temperatures (< 100°C) is a common experience. It gets worse if the surface remains contaminated for extended periods. For example, a surface may show initial tritium contamination of 10^4 dpm per 100 cm^2. Following cleaning with water, soap or detergent, the level may drop to 10^2. The next day it may go up to 10^3–10^4. Repeated washings may slowly drop it to acceptable levels of < 10^2. Heating can accelerate the removal of loosely bound tritium but may also drive the tritium deeper into the material. Bulk tritium is difficult to drive out even with prolonged heating at high temperatures. In

organic materials like rubber, plastics, etc., tritium will eventually diffuse out except for those that exchanges with organically bound hydrogen and is incorporated in the organic matrix.

Ultrasonic waves produce cavitation that assists solvent action and have been found to be effective in removing surface contamination. An aqueous solution of detergents or surface active agents is usually used as the solvent. However, it does not effectively remove tritium contamination diffused into the bulk nor does it prevent the regrowth of tritium on the surface.

Since the early days when tritium was handled, it was known that in addition to gas sweeping and application of heat, the use of moisture was also valuable in promoting isotopic exchange and removing HTO contamination. It has also been found that the use of moisture to pre-saturate a surface can decrease the adsorption of airborne HTO whereas a dry surface will readily adsorb HTO. This phenomenon is readily apparent to anyone who uses a glovebox with a dry, inert atmosphere for handling tritium. HTO can be formed in the glove box from ever-present H_2O and O_2, even at ppm levels, and adsorbs on the glove box walls. Subsequent introduction of H_2O in the box atmosphere will release this HTO although complete decontamination of the HTO in this manner is slow. In a similar manner, HT or tritiated oil adsorbed on surfaces can be removed, albeit not completely, by the introduction of H_2 or organic solvents to promote isotopic exchange [8].

Where direct heating is not possible or practical, steam cleaning has been found useful especially for large objects. Highly contaminated Van de Graaff accelerator tubes that needed to be returned to the vendor had been successfully decontaminated in this manner.

One of the earliest detailed studies on tritium decontamination was reported by Symonds [9] in which he described the decontamination of metal planchets previously contaminated by HTO. He found that heating at 300°C for 30 min or longer in air or vacuum was better than 90% effective. The effectiveness depends on the type of metal (brass was found to be better than steel which in turn was better than aluminum), and on the level of original contamination (the higher the level, the more difficult the decontamination). He also described that quenching the heated object in hot water (or better: hot water with detergent) removed much of the remaining tritium. He found contamination regrowth if the baking was done at 100°C but not at 300°C.

Boutot and Schipfer [10,11] carried out systematic studies on tritium decontamination by four general methods: (a) wet chemical treatment, (b) vacuum treatment, (c) air sweeping, and (d) electrolysis. Wet treatment tests indicated that about 95% of the activity was removed by heating carbon steel components in water at 100°C for 30 h and the result was slightly improved with the same treatment time when 1N NaOH was used. With stainless steel specimens, about 95% re-
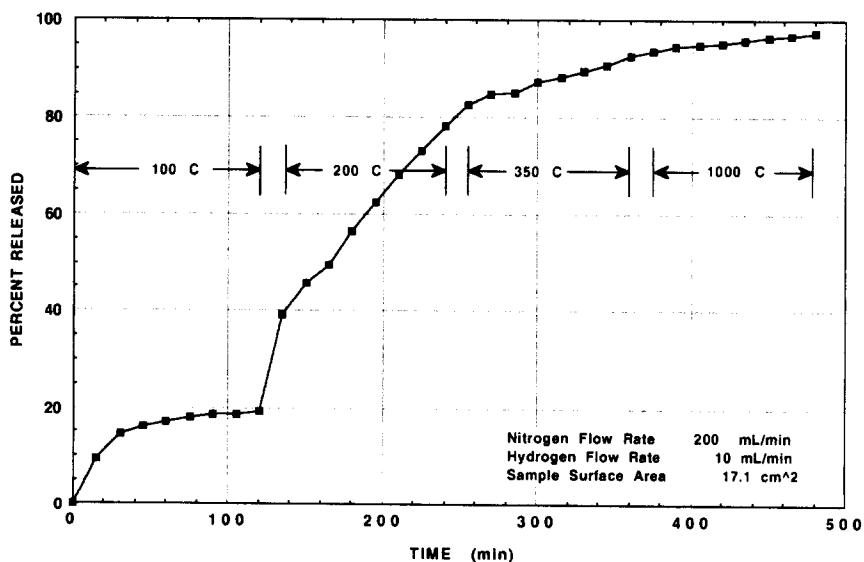

Fig. 1. HTO Release from stainless steel sample.

moval was obtained with 1N HNO_3 at 80°C after 60 h. The addition of 90 g/L of H_2O_2 increased the amount removed marginally to 97%. Similarly, vacuum treatment was shown to be a rather slow process. About 99% removal was obtained for carbon steel components after heating for 30 h at 250°C or 25 h at 350°C under 4×10^{-3} Torr. Decontamination by air sweeping was found to be very slow at 100°C. However, the removal after 60 h increased from 35% to 97% when the operating temperature was increased from 100 to 250°C. The effectiveness could be improved by adding water vapour to the air stream but not by increasing the air flow rate. The authors recommended air sweeping with or without water vapour and wet chemical processes. Tritium regrowth was experienced during storage. The extent of the regrowth depends on the initial level of contamination, the nature of the treatment used and the duration of storage.

A section of contaminated process piping from the Darlington Tritium Removal Facility, Canada was removed for decontamination study [12]. Thermal desorption of HTO and HT is shown in figs. 1 and 2. The predominant species released from the surface was HTO (> 95%). The ratio HTO/HT released depends, amongst other things, on the heating rate. Only 20% of the tritium was released at 100°C. Heating to 350°C removed 70% of the tritium within the first hour and 92% of the tritium after 2 h. Heating to 1000°C removed essentially all the tritium in a very short time

(about 1 min). It was concluded that thermal desorption at 350°C was effective since this temperature is not expected to adversely affect the ability to weld the piping.

Studies of thermal desorption of HT exposed stainless steel [13] showed a broad peak at 540 K corresponding to HTO release and three HT peaks at 430, 750 and 970 K. HTO comprised about 90% of the total tritium released. The 430 K HT peak corresponds to HT sorbed primarily on the surface. The higher temperature HT peaks probably correspond to atomic tritium which diffused into the bulk and held in microstructures.

A basic study on the tritium outgassing from highly tritium-contaminated type 316L stainless steel where tritium had permeated through the bulk and was not just confined to the surface is underway at Bruyères-le-Châtel, CEA [14]. Several samples were loaded with tritium under pure tritium pressure (25 bar) at 450°C which produced an initial tritium content of about 140 mm^3/g. The high pressure and temperature used ensured that tritium permeated through the bulk. After aging at low temperature (to avoid tritium desorption) to generate ^3He in situ, the samples were used for welding tests. Before and after the welding tests, thermal desorption studies were carried out to establish optimal conditions for obtaining acceptable residual tritium levels. Experiments at 450°C for 300 h under argon flow showed that the surface contamination was
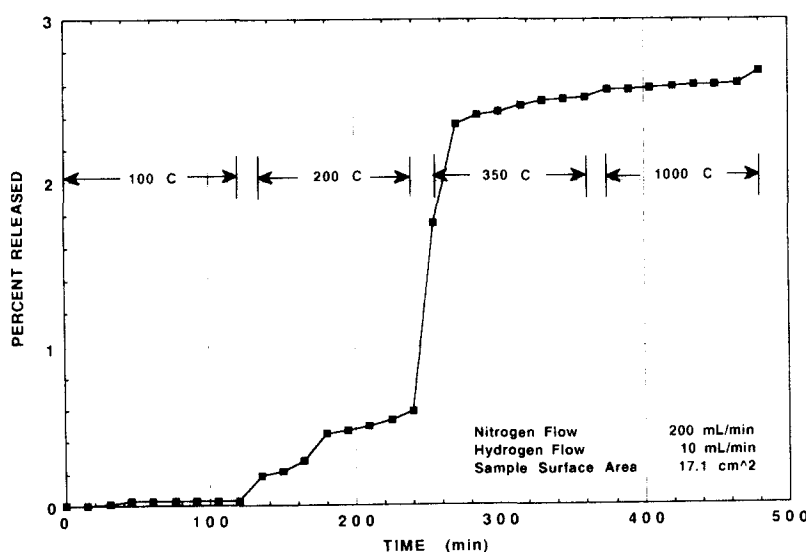

Fig. 2. HT Release from stainless steel sample.

reduced from an estimated initial level of about 1 GBq/m² to 2 MBq/m², a reduction factor of 500. The distribution of tritium through the bulk of the sample following decontamination is being studied.

In practice, a combination of washing, purging, thermal desorption, and isotopic exchange is the most commonly used method of tritium decontamination. Although it generally does not provide complete decontamination, it is often adequate to permit equipment maintenance to be safely carried out or allow components to be released from controlled areas. The following examples illustrate application of this method in practice.

A small turbomolecular pump at Ontario Hydro Research Division's tritium laboratory (OHRD) required bearing replacement. In preparation for this task, the pump oil was drained and immobilized. A fresh charge of oil was circulated through the pump for approximately five minutes and also disposed. Surface activities at the pump inlet were about 2.2 MCi/m². Detritiation of the pump consisted of heating the entire pump to 100°C and purging the desorbed tritium with an argon stream. Hydrogen and water vapour were added periodically in the purge stream to enhance tritium desorption by isotopic exchange. Tritium in the effluent was monitored with an ionization chamber during the decontamination process which spanned 510 h. Progress during the first 200 h is shown in fig. 3. The initial purge gas was argon containing 8% H_2.

Tritium concentration in the effluent dropped by a factor of 20 during the first 70 h. Then the purge gas was changed to humid argon which released substantial amounts of tritium. Purging with a dry argon stream during the period 98–122 h showed that the effluent activity had reached a steady state. Further addition of H_2 only had a moderate effect. However, a second addition of moisture at 150 h released some more tritium. Surface activities at the pump inlet at the end of the decontamination were about 37 kCi/m², i.e. a reduction factor of 60 was achieved.

A common experience in a tritium laboratory is contamination of tritium monitors. At OHRD, a contaminated stainless steel ionization chamber showed background tritium readings of about 37 MBq/m³ (1 mCi/m³). It was decontaminated by baking at 330°C for five hours. Figure 4 showed tritium concentrations of the gas leaving the monitor. Tritium desorption increased with increasing temperature and subsequently decreased as the loosely bound tritium was freed from the surface. Injection of air, with a relative humidity of about 30%, enhanced tritium desorption from the surface.

3.2. Etching

Chemical or electrochemical etching decontaminates by removing a very thin layer of surface material, in particular the oxide layer where the tritium is chemi-
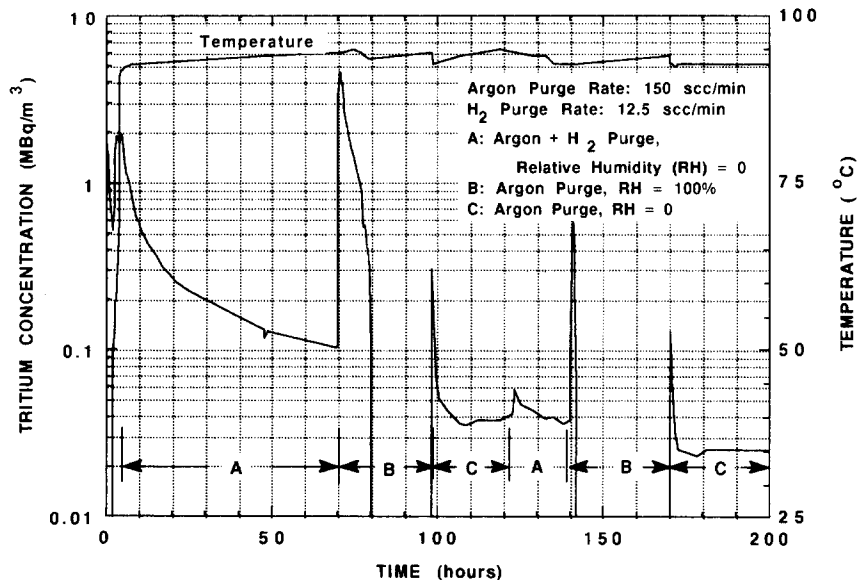

Fig. 3. Tritium decontamination of aluminum pump casing.

cally bound. Generally the objective is to remove less than 1 μm of material. This method generates mixed chemical and radioactive wastes. Also, the surface may be slightly roughened by the treatment which facilitates re-contamination if the component is re-used. Nevertheless, the method can be highly effective for some applications.

Hirabayashi et al. [15–17] studied in detail surface sorption of HT gas, thermal desorption and decontamination by chemical etching of type 316 stainless steel. Etching with dilute acids [15] removed the thin surface oxide film in which tritium was trapped. The major part of the tritium released from the surface was in the HTO form which was present within about 0.01 μm from the surface. The HTO was released quickly in the initial stage of etching while the release of HT lasted for a long time. Release of tritium upon soaking in water is slow with about 90% of sorbed tritium released as HTO within one day but 10% still remaining after 5 d.

A combination of chemical and thermal methods were proposed for the detritiation of type 316 stainless steel components [14,17]. For components which sorb tritium only on the surface, a two-step process was found to remove more than 95% of the tritium. The specimen was first dipped for 2 h in 5% HCl for whole surface corrosion, then for 24 h in 5.5% $CuSO_4$–15.7% H_2SO_4 solution for intergranular corrosion. Dissolution of material was minimal (2.45 mg/cm^2). For prac-

tical specimens in which tritium had also permeated into the bulk, the specimen was first vacuum-baked for 24 h at 773 K, followed by etching for 2 h in dilute HCl. This reduced tritium contamination by a factor of about 200.

Chemical etching following water leaching, using a dilute HNO_3–HF solution, was used successfully to decontaminate the inside of a gas bottle used to store tritium [18]. The decontamination was carried out prior to welding a replacement valve on the gas bottle.

Electrolysis using various 10 g/L acids and bases such as sulphuric acid, perchloric acid, phosphoric acid and sodium hydroxide produced very high tritium decontamination factors at 25°C in a relatively short time [10]. For example, in sulphuric acid, at least 99% of the activity was removed from carbon steel components. However, because of corrosion problems, the method is not suitable for the decontamination and reuse of precision-machined components although it could substantially reduce the volume of tritiated waste for disposal and offer the possibility of recycling equipment for the scrap metal market.

Bellanger [19] described an electrolysis-based tritium decontamination method which minimized erosion loss of surface material and is applicable to small metal parts with complex geometry. The contaminated part is connected to the negative pole of a direct current generator. An aqueous electrolyte able to release hydrogen by electrolysis is used. Low electrolysis
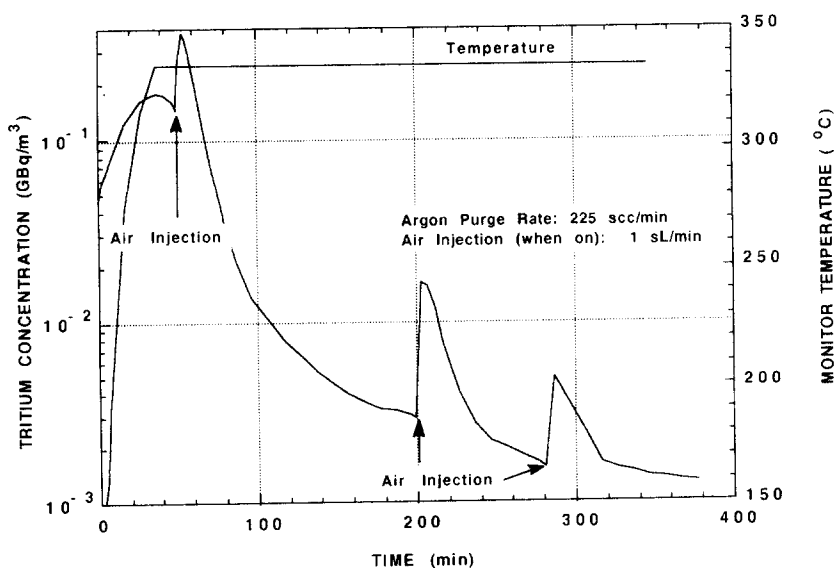

Fig. 4. Decontamination of stainless steel tritium monitor.

current densities (10–50 mA/cm²) are used to cathodically charge the contaminated surface with hydrogen thereby replacing tritium adsorbed on the surface with hydrogen. The low current densities used minimize surface material erosion. In one example cited, 1N NaOH was used as the electrolyte. Electrolysis was carried out over a 2 h period at 80°C and 10 mA/cm². The decontamination ratio (activity before/activity after treatment) was 10 after one cycle and the surface material loss was 10.2 μm. After 12 cycles the decontamination ratio was 10^4.

The electrochemical behaviour of type 304L stainless steel was studied by cyclic voltammetry in 0.1 N NaOH [20]. The hydrogen produced by electrolysis at electropotential values more negative than −1.6V diffused into the electrode; at more positive potentials, it diffused back towards the surface where is it reoxidized. Sheets contaminated by tritium were studied on the basis of the above results. At −0.3V, the oxide layer is not destroyed and the tritium contained in it is oxidized and rapidly passes into the solution. Smaller amounts of tritium diffusing from the bulk are also subsequently oxidized. At −1.3V, essentially the same phenomenon is observed except that the oxide layer is destroyed. At −1.6V, penetration and oxidation of the tritium occur at the same time.

3.3. Plasma discharge cleaning

Glow discharges have been used for many years in the cleaning of vacuum systems and is used in the cleaning and conditioning of tokamaks, for example at JET and TFTR. The plasma energy can be much higher than that achieved thermally and yet will not damage surfaces because of the low thermal flux. This is a significant advantage over thermal desorption methods of decontamination which take a long time and may be limited by the temperature limits of materials. The bonding energy of the water molecule to metal surfaces is high, typically 1.5 eV or more [21]. This energy level is much higher than the melting point of iron and hence thermal desorption is slow. Plasma cleaning has the potential advantages of simplicity, low cost and adaptability to different sizes and shapes of surfaces.

Detritiation of metallic components by immersion in a low pressure plasma is being studied at Ontario Hydro's Research Division, Canada [22]. Tritiated items are placed in the plasma reaction chamber and desorbed species are continuously swept from the reaction zone. The performance of the system has been studied for a range of discharge gases and pressure conditions.

Table 1
Tritium decontamination of stainless steel components by plasma discharge

Working gas	Surface activity (dpm/cm²)	
	Initial	Final
Hydrogen	13629	4759
	4759	3198
	3198	2530
Helium	13936	942
Argon	18898	889

Reduction of surface tritium contamination may be achieved within a few minutes. Some preliminary results are given in table 1. The surface did not warm up appreciably (< 40°C) showing that decontamination was achieved without surface heating. Preliminary results suggest that tritium regrowth on the surface is slow.

While the results of metal decontamination using a low pressure plasma are very encouraging, it is probably too early to implement a facility in the field. Several questions need to be resolved. Which gas provides the best decontamination results? What is the optimal operating pressure range? Higher pressures are better from a facility design standpoint since the complexity of the vacuum system can be dramatically reduced. How can contamination of the plasma chamber be minimized so that cross contamination of components being decontaminated can be reduced? What is the tritium regrowth rate? Can inner surfaces and complex geometries be decontaminated effectively? With proper adaptation, the method may be used to decontaminate the inside walls of metal-clad caissons or contaminated equipment transport flasks or casks.

In graphite-lined tokamak reactors, codeposition of eroded carbon with tritium will result in large inventories of tritium in the torus. If there is an accidental loss of vacuum for the torus resulting in air entering the vacuum vessel, much of the tritium could be released. Experiments [23] performed in the Tritium Plasma Experiment (TPX) at Sandia National Laboratories in Livermore (SNLL) showed that a sizeable fraction of the tritium in a thin (20 nm) codeposited film was released upon exposure to air. Heating the film to 573 K caused complete release of the tritium. The tritium was released primarily as HTO.

A glow-discharge technique has been proposed for removing this film periodically to reduce the inventory buildup [24]. This technique, which was tested in the Laser Material and Plasma Experiment (LAMPE) at SNLL, utilizes a He/O_2 glow-discharge to volatilize

the carbon deposit and liberate the codeposited tritium. The LAMPE studies showed the carbon film could be etched at a rate of about 0.1 nm/s. In the case of the Burning Plasma Experiment (BPX), it is envisioned that this removal process needs to be applied once after each full week of normal tritium plasma operation. Each cleaning cycle will last for about 24 h to remove the estimated 1 g of tritium that has been accumulated.

3.4. Destructive decontamination methods

Highly tritiated components that have to be disposed as waste may be detritiated and volume-reduced by destructive techniques. Tritium recovery may also be justified on economic grounds. Thermal desorption is first carried out under an inert atmosphere. The outgassed tritium is recovered in a detritiation unit. Then the material is melted at high temperature to recover the bulk of the remaining tritium and to produce a volume-reduced waste form in which the residual tritium corresponds to the limit of solubility at the melting temperature.

In 1983, about 4 tonnes of metallic wastes had been processed at Valduc, CEA [25]. Initial thermal desorption was carried out at 800°C. The mean tritium activities were 19 MBq/g and 0.15 MBq/g before and after treatment. After melting of the materials at 1400 °C under vacuum, the levels of mean activity were variable (< 37 kBq/g, or 1 Ci/t) and the surface activity corresponded to 7 Bq/g. The resulting ingot was safety manipulated out of the glove box and is below the limit of 2 Ci/t for shallow land burial in France.

Graphite tiles used for first-wall protection which contains both tritium and some ^{14}C could be processed in the same furnace used to recover tritium by melting steel [26]. At the operating temperature of about 1500°C, most of the tritium or tritiated hydrocarbons will outgas from graphite and carbon will be immobilized in the steel.

4. Decontamination at tritium removal facilities

4.1. Institut Laue–Langevin (Grenoble) tritium extraction facility

The tritium extraction facility at Grenoble, France, is the first major facility extracting tritium from heavy water. Starting operation in 1971, it has extracted more than 7.2×10^4 TBq (2×10^6 Ci) of tritium by 1988 [27]. Welding on tritium-contaminated pipework has been performed on several occasions. The following procedure has been consistently used to decontaminate the piping prior to welding:
- pump down,
- purge with clean D_2 (the number of purges depends on the circuit where the work is being done),
- fill with helium before the circuit is opened,
- isolate the circuit from the remainder of the facility by solid flanges or a double barrier filled with inert gas,
- protect workers with ventilated masks specially adapted for welding.

4.2. Darlington tritium removal facility

The Darlington Tritium Removal Facility (DTRF) in Ontario, Canada started operation in 1988. It is presently the world's largest tritium extraction plant, recovering tritium at a rate of about 2×10^4 TBq (6×10^5 Ci) per week from heavy water with an average concentration of 0.37 TBq/kg (10 Ci/kg). Recent modifications to the high tritium cryogenic distillation system required cutting and rewelding of highly contaminated process piping. Tritium decontamination experience was described by Guglielmi et al. [28] and by Carmichael and Haynes [29]. The tritium gas concentration in the system was estimated to be about 2×10^3 TBq/m^3 at ambient temperatures. The measured surface contamination level inside the piping was about 0.1 TBq/m^2 (3 Ci/m^2). Most of this would be expected to be released instantly when welding started.

The process system was evacuated and backfilled with hydrogen every 3–4 d for approximately one month to help promote isotopic exchange with tritium and reduce the formation of tritium oxide. System layout would not allow recirculation or heating of the hydrogen during the soaking period. The piping was then evacuated and backfilled with helium to provide an inert atmosphere for worker protection during welding.

A ventilated, double plastic wall enclosure was established around the contaminated work area. The cold box was disassembled and the piping cut. Maximum tritium oxide concentrations during pipe cutting were 3.7 MBq/m^3. The cut area was then heated with a propane torch to drive off the bulk of the tritium. Tritium oxide levels during heating reached about 3.7 GBq/m^3 at 10 cm from the open tube ends, about 37 MBq/m^3 at 30 cm away and about 3.7 MBq/m^3 in the work area of the enclosure. Welding of the replacement piece was then carried out under argon purge. 'Elephant trunk' local ventilation was provided directly above the workers who were also protected in double

plastic suits. The general room tritium concentrations were less than 0.37 MBq/m^3 (10 μCi/m^3) and the concentrations directly above the weld ranged between 37 MBq–1 GBq/m^3. Pre-heating with the propane torch appeared to be effective, and resulted in relatively low hazard levels. Surface tritium contamination of the outer plastic suit was significant. It was found that wetting of the inner plastic suit for 3–5 min before removal was an important step to reduce personal tritium surface contamination to insignificant levels.

A special enclosure called the 'A box', a dead-end leg of the high tritium cold box built for sampling, was exposed to a tritium gas leak. Tritium surface contamination inside the box was greater than 0.3 TBq/m^2. Heating the box to 450°C with inductive heating bands for 6 h reduced the contamination level by a factor of 10–20. The room surface contamination levels were reduced by humidifying and heating the enclosure for 24–48 h as well as direct steam cleaning of the floor. Surface tritium contamination was reduced by a factor of up to 200 for smooth steel or plastic surfaces but only by a factor of about 10 for rough, porous surfaces.

5. JET remote handling tools decontamination studies

Experiments [30] were performed at TSTA (Tritium Systems Test Assembly), Los Alamos National Laboratory to determine the potential tritium contamination of remote cutting and welding tools to be used at JET (Joint European Torus) following introduction of DT fuel, and to evaluate the relative efficacy of decontamination techniques. Pipe specimens were exposed to HT at 4.6×10^{10} Bq/m^3 for 18–168 h and then cut or welded by the remote tools. A maximum tritium release of 1.6×10^4 Bq was measured during the welding, resulting in minor contamination of the tools (0.5 Bq/cm^2).

Following these tests, planchets of stainless steel, aluminum and rigid PVC were exposed to HTO liquid at a concentration of 4.4×10^{10} Bq/L for 1, 24, 120 h and decontaminated. The decontamination techniques used included (a) 24 h outgassing at room temperature, (b) 24 h leaching in water at room temperature, (c) 24 h baking at 100°C, (d) 1 h heating in a hot air stream at 125°C followed by 23 h outgassing in room air. The maximum levels of tritium surface contamination measured during the test, before decontamination, were approximately 12 Bq/cm^2 for stainless steel, 5 Bq/cm^2 for aluminum alloy and 1700 Bq/cm^2 for PVC. About 80% of the surface activity was removed using a hot air

stream at 125°C on stainless steel and aluminum alloy and baking PVC at 100°C.

6. Tritium contamination of walls

Outgassing of tritium that has diffused into containment walls gives rise to chronic airborne tritium contamination. Following an acute tritium release into containment, the time taken by atmospheric detritiation systems to reduce air tritium concentrations to acceptable levels can be greatly increased due to outgassing of tritium sorbed in the containment walls.

Most surfaces absorb more HTO than HT. The degree of tritium absorption on various surfaces had been measured and ranked below [31]. Samples measured were as received, i.e. there was no surface treatment other than simple degreasing. Surface polishing has been found to decrease the amount of HT absorbed by a factor of about 5 and some decrease of HTO absorption has also been observed.

For HTO:

concrete, gypsum board ≻ paint, vinyl tile, porcelain tile > metals > polished metals, glass.

For HT:

concrete > vinyl tile > oil-base paint > porcelain tile > gypsum board > metals > glass.

Within the metals:

galvanized steel > brass > SS-304 steel > copper > mild steel > aluminum > tin-plated steel > polished SS-304 steel.

In another study, Sienkiewicz [32] found the following tritium absorption ranking for materials exposed to DT gas:

stone block > copper tubing > plexiglass > stainless steel > aluminum.

Correlations between atmospheric tritium concentration and surface tritium contamination have been established for HTO and HT, for stainless steel surfaces [33]. Options for decontamination of metallic walls are washing, air/gas sweeping with or without moisture/H$_2$ addition, and plasma discharge cleaning.

Bare concrete surfaces are undesirable because they are porous and readily absorb tritium, especially HTO. Decontaminating the concrete wall itself by air purging is very slow. The addition of moisture should help but will generate more liquid waste. If the tritium has not significantly penetrated the concrete, vacuuming the

Table 2

Characteristics of wall protective coatings

Performance	Organic-based coatings	Metallic coverings	Ceramic coatings or coverings
Tritium absorption and outgassing	poor	good	very good
Tritium permeation resistance	poor	good	very good
Minimization of HT to HTO conversion	good	fair	good
Resistance to damage	good	very good	fair
Ease of decontamination	fair	good	very good
Cost	low	moderate	moderate to high

surface and then scrubbing with water and detergents or solvents may help. More drastic measures such as water cannon and concrete spaller, though not generally recommended, have been described [34].

Tritium absorption in concrete that has been chronically exposed to HTO contaminated atmosphere was studied by Krasznai [35]. Preliminary data showed that the rate of desorption of HTO is diffusion controlled. The effective diffusivity of HTO in concrete is about 4 orders of magnitude lower than the self diffusion coefficient of tritiated water in light water. Most of the tritium appears to be confined within the first 10–20 cm of the surface.

It would be best to avoid wall contamination in the first place by minimizing chronic airborne tritium, particularly HTO concentrations, by reducing the contact time following acute releases into containment, and by applying a protective coating over the porous surface of concrete. Ideally, such a coating should have low absorption/outgassing and low permeation rate for both HTO and HT, and should be easy to apply forming a tight, chemically compatible bond with the concrete surface. For exposure to HT gas, it would also be desirable that the coating does not promote conversion of HT to HTO.

Work was undertaken in 1982 and in 1985 at TSTA [36,37] to evaluate tritium retention and outgassing by wall coatings. Epoxy and latex paints were tested as well as aluminum foil. HT gas did not significantly contaminate the chamber walls. The amount of HT converted to HTO was a small fraction of the total: 10^{-5}–10^{-4} with more conversion in the chamber lined with aluminum foil. Latex paint initially absorbed more HTO but was found to release it more readily than epoxy paint. However, epoxy has more favourable physical properties such as hardness and durability. Aluminum surfaces were found to be less likely to become significantly contaminated.

Braun and Williamson [38] studied tritium retention/outgassing and permeation for 3 types of coat-

ings: epoxy, vinyl and urethane. The data suggested that HTO is desorbed faster from the more open film structure found in coatings with high pigment-to-binder ratios, such as epoxy. Desorption of HTO was more effective at higher humidities (60% RH at room temperature) and elevated temperature (60°C). HTO and HT were found to permeate through the coatings at comparable rates; the rates were somewhat lower for epoxy coatings.

Ono et al. [39] studied the sorption and desorption behaviour of HTO in epoxy, acryl and silicone based coatings. The acryl paint exhibited a smaller retention of HTO compared to the others. The epoxy was found to have a higher desorption rate than the others but the fraction of tritium not recovered by purging at room temperature was also higher.

Table 2 provides a comparison of the performance characteristics of different surfaces. Organic-based coatings can be easily and inexpensively applied and are an improvement over the bare concrete surface. They still have significant disadvantages especially high tritium absorption/outgassing and permeation. The use of strippable paints may be considered. Metallic coverings are substantially better than organic-based coatings if they can be applied without gaps or seams through which tritium can penetrate. They can also be expensive if required to cover the walls of large containment buildings. Ceramic, enamel or glass-like coatings or coverings would be best if they can be easily and economically applied to concrete surfaces. They show the lowest tritium absorption and outgassing and are effective tritium permeation barriers. Further R & D on the development and characterization of such coatings will be highly desirable.

7. Tritium decontamination considerations at fusion facilities

For a D–T tokamak device, systems that are subject to tritium contamination are:

(a) Systems directly associated with tokamak operation
These include the torus vessel, first-wall, divertor, in-vessel shield, breeding blanket, fuelling, primary cooling, vacuum pumping, neutral beam injectors, RF heating, and plasma diagnostics. They are mainly semi-permanent components. Removable components are basi-cally the in-vessel items such as the divertor plates, first-wall tiles, and blanket modules. They are subject to both neutron activation and tritium contamination. The in-vessel components are also subject to activated erosion dust contamination. Radiological hazards during maintenance are dominated by gamma radiation
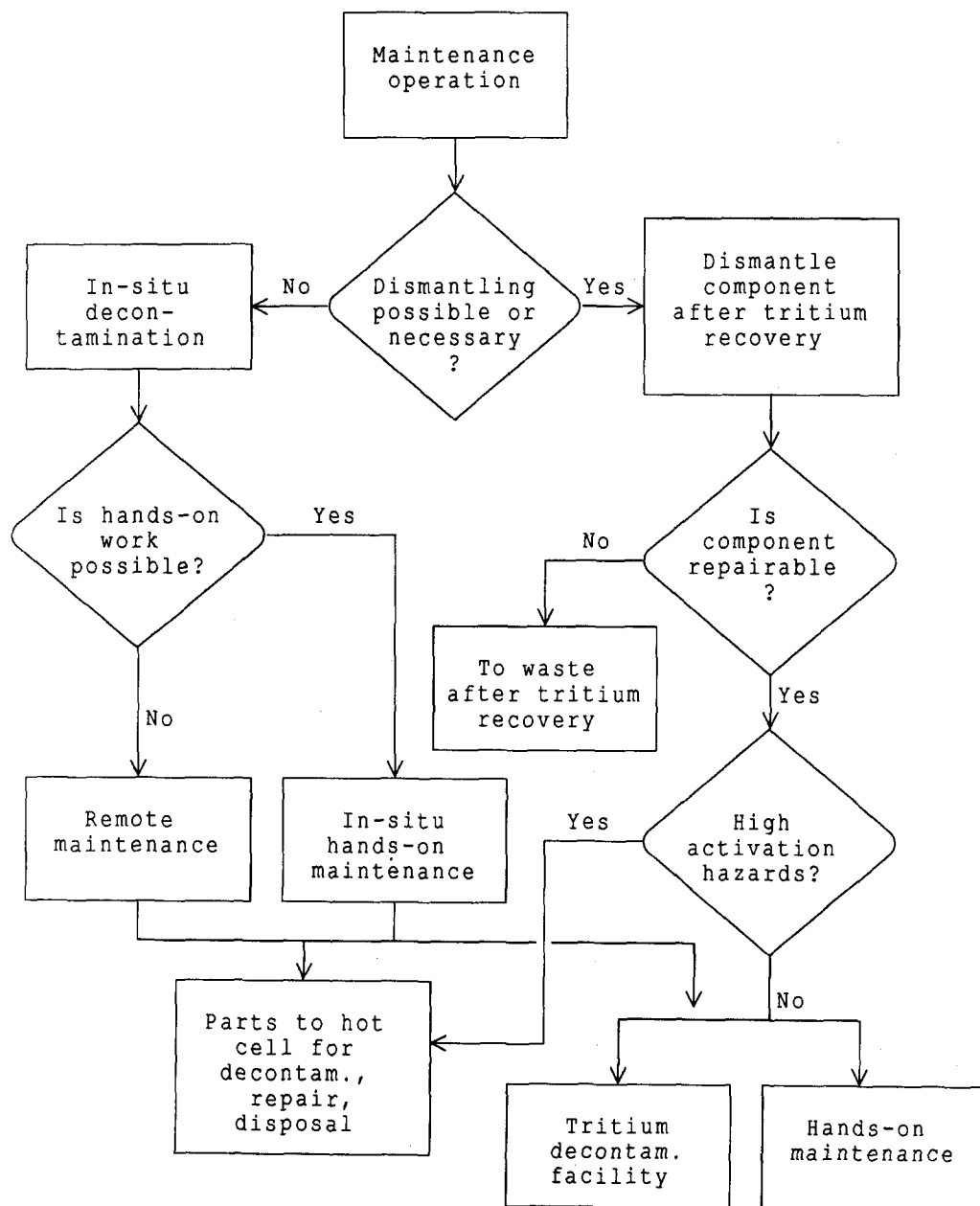

Fig. 5. Tritium decontamination considerations.

and hence require remote handling. In-situ tritium decontamination, if required, will be more difficult.

(b) Tritium processing and handling systems

These include the plasma exhaust cleanup, hydrogen isotope separation, blanket tritium extraction, tritium storage, atmosphere and water detritiation systems. High to very high levels of tritium contamination may be present in these systems. With the exception of small amounts of activated dust in the plasma exhaust cleanup and atmospheric cleanup systems, there is little gamma hazard.

(c) Maintenance and waste management systems

These include in-vessel and ex-vessel remote maintenance equipment, contaminated equipment transfer flasks or casks, hot cells, waste immobilization, conditioning and handling equipment.

(d) Radioactivity containment systems

These include containment walls, penetration seals, access structures and active ventilation systems.

Tritium decontamination considerations are summarized in fig. 5. Some components are fixed or semi-permanent. Decontamination of these components or process piping may have to be carried out in-situ prior to cutting and rewelding operations. This is particularly important if hands-on maintenance is required. Other components are removable and may be transported to the hot cell or to dedicated tritiated equipment maintenance facilities for decontamination and repair.

If the component is not removable and tritium decontamination in-situ is judged to be necessary or desirable before maintenance activities, the system can be vacuum pumped or swept with a purge gas with added H_2O and/or H_2. Heating, if feasible, will speed up the process. For stainless steel piping, heating to about 350°C has been found to be effective without adversely affecting welding.

Most of the tritium processing systems where there is little or no activation product hazard permit hands-on maintenance. Experience at the Grenoble and Darlington tritium removal plants showed that in-situ, hands-on maintenance can be safely carried out even on very highly tritiated systems, following tritium decontamination and with proper radiation protection procedures and equipment.

For components that are removable, it is still good practice to carry out in-situ decontamination prior to removal. This will recover much of the tritium and will minimize tritium contamination spread and subsequent
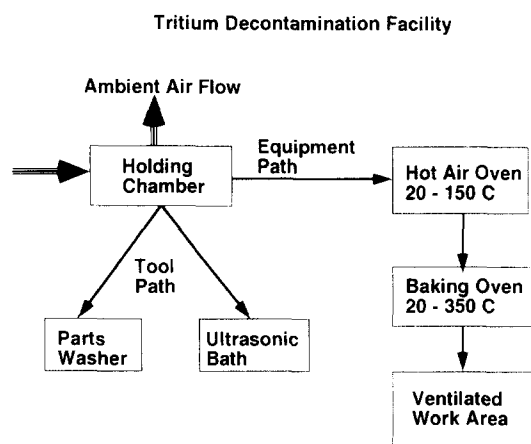

Fig. 6. Tritium decontamination facility.

handling problems. For graphite-lined tokamak reactors, glow discharge cleaning is periodically carried out to maintain a clean surface and to reduce tritium inventory in the graphite.

A dedicated tritium decontamination facility may be provided for components that have little or no activation product hazard. The absence of external gamma radiation permits human access and hands-on work. It may be located in the Tritium Building, where it could be readily accessed from tritium processing equipment rooms. A conceptual outline of such a facility is shown in fig. 6. A ventilated holding chamber allows storage and outgassing of components. Some parts may be decontaminated using an industrial dishwasher or ultrasonic bath. A hot air oven operating up to 150°C is used to provide first stage thermal desorption decontamination. There is provision for adding moisture during thermal desorption. A second stage thermal desorption is done in a baking oven operating up to 350°C. Separating thermal desorption into two stages minimizes cross-contamination of components; the second stage is the finishing stage where residual tritium after the first stage is removed. Other equipment may be included such as a plasma discharge cleaning chamber or special chemical or electrochemical detritiation facilities.

Tritium decontamination of components in the hot cell, if required, is designed to be carried out remotely. Storage and outgassing, preferably at elevated temperatures (< 100°C), are the simplest methods. Water jets or steam jets may be used for external decontamination of large components. There should be provision for purging and vacuuming of the inside of components. For small parts, remotely operated washing ma-

chines or ultrasonic baths may be used. A plasma discharge cleaning chamber may also be operated remotely.

Recovery of tritium from components such as carbon tiles and destructive decontamination of structural components by melting are also carried out in the hot cell.

8. Conclusions

Tritium contamination is characterized by the high mobility of tritium resulting in ready cross-contamination, absorption and permeation in many materials. Tritium outgassed from surfaces is primarily in the HTO form. The most commonly used methods of tritium decontamination are washing, vacuuming, purging, thermal desorption, and isotopic exchange by adding H_2O and/or H_2 in the purge gas. These are generally adequate but they are tedious and time consuming. Experience at large tritium removal plants has shown that it is possible to decontaminate in-situ highly tritiated process components to permit hands-on work to be safely carried out with proper radiation protection procedures and equipment.

More sophisticated methods such as chemical/ electrochemical etching and plasma discharge cleaning can potentially provide high decontamination factors in a relatively short time. Chemical methods tend to produce 'mixed' wastes, i.e. both chemical and radiological wastes. They may be suitable for special small items where very high decontamination factors are desirable. Plasma discharge cleaning potentially can be attractive. It is simple, efficient, and can be applied to different sizes and shapes and warrants more R&D.

Destructive decontamination by melting has been shown to be feasible to decontaminate highly tritiated steel components to a level acceptable for shallow land burial disposal.

Tritium contamination of walls, especially concrete, presents a chronic outgassing problem. Concrete decontamination is very difficult and slow. It would be best to avoid or minimize contamination in the first place by ensuring chronic tritium concentrations are low and by covering the surface with protective coatings. Ceramic coatings seem to have the most desirable characteristics; more R&D is needed.

At DT-burning fusion reactor facilities, a dedicated tritium decontamination facility should be provided. This is specially designed for the decontamination and maintenance of components that have only tritium contamination and allows hands-on work in the absence of gamma hazard.

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